

The probability density function for the Havriliak-Negami relaxation

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We study the functions related to the Havriliak-Negami frequency relaxation $\sim [1 + (i\omega\tau_0)^\alpha]^{-\beta}$ with τ_0 characteristic time, measured in many experiments. We furnish exact and explicit expression for the response function $f_{\alpha,\beta}(t/\tau_0)$ in time domain and a probability density $g_{\alpha,\beta}(u)$ in space domain for $\alpha = l/k < 1$ and $\beta < k/l$, with k and l positive integers. For $0 < \alpha < 1$ and $\beta = 1$ we reproduce the functions related to the Cole-Cole relaxation. We use the method of integral transforms. We show that $f_{\alpha,\beta}(t/\tau_0)$ with $\beta = (2 - q)/(q - 1)$ and $\tau_0 = (q - 1)^{1/\alpha}$, $1 < q < 2$, goes over to the one-sided Lévy stable distribution when q tends to one. Moreover, applying the self-similar property of $g_{\alpha,\beta}(u)$ we introduce two-variable density which satisfies the integral form of evolution equation.

Keywords: the Havriliak-Negami relaxation, the Prabhakar function, the (three-parameter) generalized Mittag-Leffler functions

I. INTRODUCTION

In many glass-forming system, like amorphous polymers or supercooled liquids near the glass transition temperature, the relaxation spectrum exhibits strongly non-exponential behavior. Dielectric spectroscopy shows evidence of two relaxation processes: the so-called α - and β -relaxation. The asymmetric α -relaxation peak flattens into an β -relaxation at high frequency domain [1]. In general, the α -relaxation corresponds to the atomic motion of the clusters of atoms themselves or atomic transport between clusters, while the β -relaxation corresponds to the motions within the clusters. In a phenomenological approach these two types of relaxation are usually described by the sum of empirical, non-Debye laws, namely the sum of Havriliak-Negami (HN) [3] relaxation functions [4–8] or their sum with the stretched exponentials [9–11]. The stretched exponentials (named also the Kohlrausch-Williams-Watts (KWW) functions) are extensively studied in many theoretical papers, see e.g. [12–15], and experimental papers, see e.g. [16–20]. This behavior of the dielectric function can be also described by the excess wing [1, 2].

This paper is devoted to the HN function which was

introduced in [3]:

$$\frac{\hat{\varepsilon}(\omega) - \varepsilon_\infty}{\varepsilon - \varepsilon_\infty} = \frac{1}{[1 + (i\omega\tau_0)^\alpha]^\beta}, \quad 0 < \alpha < 1, \quad \beta > 0. \quad (1)$$

Here, the parameters α and β do not denote the α - and β -relaxation. They are called the width and symmetry parameters, respectively. The symbol ω denotes the frequency and τ_0 is an effective time constant. The symbols ε and ε_∞ denote the relative permittivity and the dielectric constant respectively. Note that Eq. (1) generalizes the Cole-Cole (CC) [21] relaxation (Eq. (1) for $\beta = 1$) and the Cole-Davidson (CD) [21] relaxation (Eq. (1) for $\alpha = 1$). Among experimental papers devoted to the HN functions we mention Ref. [20] where the HN function has been measured during the monitoring of contamination in sandstone. The HN relaxation is also observed in a complex system representing plant tissues of fresh fruits and vegetables in the frequency range $10^7 - 1.8 \times 10^9$ Hz, see [22]. It can be also applied to model a thermal flux in the field of machining by turning, in the time domain [23].

Several authors have investigated the HN relaxation from different points of view. For example, the relation between CC relaxation and the stretched exponential is established in [24, 25]. A transparent subordination approach to anomalous diffusion processes underlying the HN relaxation has been proposed in [26, 27]. The analytic expressions in the time domain for the HN relaxation in terms of the Fox H functions are presented in [28, 29] for the real values of parameters α and β . (Remark that the Fox H functions are defined via the Mellin transform [30].) The non-rational values of α and β are obtained by

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making the numerical fitting of the HN function with the experimentally obtained data. For this reason the solutions expressed via the Fox H functions are correct but impractical because of the Fox H functions are not yet implemented in the computer algebra systems. Therefore the expression involving the Fox H functions for general parameter values are purely symbolic and, for the moment, cannot be used in actual calculations.

The purpose of this paper is to express the relevant functions, like the response functions, the probability densities and the relaxation functions, related to the HN relaxation in terms of the special functions implemented in the computer algebra systems. That can significantly simplify the numerical calculations. To realize this purpose we approach the non-rational value of parameter α by the rationals such that $0 < \alpha < 1$. Hence, everywhere in the paper we take $\alpha = l/k$ with k and l relative prime integers. Another aim of the paper is to propose the probability density related to the response function. That will hopefully allow one to find localization of the relaxed center in the given sample.

The present paper is organized as follows. Sec. II contains the basic facts about the relaxation theory, namely the relation between the response functions, the probability densities and the relaxation functions. In Sec. III, the response functions $f_{\alpha,\beta}(t/\tau_0)$, $0 < \alpha < 1$ and $\beta > 0$, are represented via the Prabhakar function which contains the (three-parameter) generalized Mittag-Leffler functions. For $\alpha = l/k < 1$, where k and l are integers, we represent the (three-parameter) generalized Mittag-Leffler functions as the finite sum of the generalized hypergeometric functions. Next, we find the asymptotic properties of the response functions and the (three-parameter) generalized Mittag-Leffler functions. By expressing the response function in terms of the Meijer G function in Sec. IV we calculate the probability density $g_{l/k,\beta}(u)$ connected with the HN relaxation. We show that for $\beta < k/l$, $g_{l/k,\beta}(u)$ is positively defined and normalized. Thereafter, we find all the moments and asymptotics of $g_{\alpha,\beta}(u)$. In Sec. V the relaxation functions appropriate to the HN relaxation denoted as $[n(t)/n_0]_{\alpha,\beta}$ are derived. There the integral form of evolution equation is also found. The new relation between $f_{\alpha,\beta}(t/\tau_0)$ and the one-sided Lévy stable densities is found in Sec. VI. The paper is concluded in Sec. VII.

II. SKETCH OF RELAXATION THEORY

In the theory of relaxation the complex frequency-dependent absolute permittivity (dielectric constant) of the material is given by [21]

$$\frac{\hat{\varepsilon}(\omega) - \varepsilon_\infty}{\varepsilon - \varepsilon_\infty} = \mathcal{L}[f(t/\tau_0); i\omega]. \quad (2)$$

The symbol $\mathcal{L}[f(t/\tau_0); p] = \int_0^\infty e^{-pt} f(t/\tau_0) dt$, $\text{Re}(p) > 0$, defines the Laplace transform of $f(t/\tau_0)$ [31]. The response function $f(t/\tau_0)$ (the number of

polarized center per unit time) is related to the ratio of polarization at time t to all possible polarization via $f(t/\tau_0) = -\frac{d}{dt} \frac{n(t)}{n_0}$. Inserting this relation into the Laplace transform in Eq. (2), comparing with Eq. (1), and using Eq. (3-4-1) of [31] we have

$$[1 + (i\omega\tau_0)^\alpha]^{-\beta} = 1 - i\omega \mathcal{L}\left[\frac{n(t)}{n_0}; i\omega\right]. \quad (3)$$

From Eq. (3), Hilfer calculates all possible relaxations (called also the relaxation functions) $n(t)/n_0$ in [28, 29] using the Fox H function and the series. His relaxation function expressed via the Fox H function can be obtained by calculating the Laplace transform in Eq. (3) and, thereafter, inverting it.

On the other hand $n(t)/n_0$ can be considered as the relaxation of the sample which contains N centers of oriented polarization. Each of these centers relaxes with a different relaxation time $\tau_k = \tau_0/u_k$. The relaxation of all sample should be the weighted sum of the Debye's relaxations. That is

$$\frac{n(t)}{n_0} = \sum_k e^{-t/\tau_k} g(u_k) \Delta u_k.$$

The probability distribution $g(u_k) \Delta u_k$ is a positive function which satisfies $\sum_k g(u_k) \Delta u_k = 1$. Taking the infinitesimally small Δu_k and going with N to infinity we get

$$\frac{n(t)}{n_0} = \int_0^\infty e^{-\frac{t}{\tau_0}u} g(u) du = \int_0^\infty e^{-v} g\left(\frac{\tau_0}{t}v\right) \frac{\tau_0}{t} dv, \quad (4)$$

where $v = \tau_0 u/t$. Taking the derivative over time of the first equality in Eq. (4) the response function reads

$$f(t) = \int_0^\infty e^{-\frac{t}{\tau_0}u} \frac{u}{\tau_0} g(u) du. \quad (5)$$

We point out that the equality of $n(t)/n_0$ derived by Hilfer (see Eq. (3) where the information about the Debye's relaxation is absent) to $n(t)/n_0$ given by Eq. (4) would mean that the HN relaxation is somehow built from the Debye's relaxation.

III. RESPONSE FUNCTION

Let us denote the response function in time domain related to the HN relaxation as $f(t/\tau_0) \equiv f_{\alpha,\beta}(t/\tau_0)$. The subscripts α and β are the width and symmetry parameters in Eq. (1). We calculate the function $f_{\alpha,\beta}(t/\tau_0)$ by comparing Eqs. (1) with (2), and, next, we invert the Laplace transform. Thus, by using Eq. (2.5) of [32] for $\beta_P = \alpha\beta$, $\rho_P = \beta$ and $\lambda_P = -1$ the response function can be expressed via the so-called Prabhakar function [32-34]:

$$f_{\alpha,\beta}\left(\frac{t}{\tau_0}\right) = \tau_0 \left(\frac{t}{\tau_0}\right)^{\alpha\beta-1} E_{\alpha,\alpha\beta}^\beta\left(-\left(\frac{t}{\tau_0}\right)^\alpha\right). \quad (6)$$

(The subscript P, as the various subscripts below, is only added to emphasize the reference from which the formula was taken.) The (three-parameter) generalized Mittag-Leffler function $E_{\alpha,\gamma}^\delta(z)$ is commonly given through the series [33–35]

$$E_{\alpha,\gamma}^\delta(z) = \sum_{n=0}^{\infty} \frac{(\delta)_n z^n}{n! \Gamma(\alpha n + \gamma)}, \quad (7)$$

where $(\delta)_n = \Gamma(\delta + n)/\Gamma(\delta)$ denotes the Pochhammer symbol. Remark that for $\delta = 1$ Eq. (7) gives $E_{\alpha,\gamma}^1(z) = E_{\alpha,\gamma}(z)$ which is the (two-parameter) Mittag-Leffler function [35]. For $\delta = \gamma = 1$ Eq. (7) is equal to the (classical) Mittag-Leffler function $E_\alpha(z)$ [35], and for $\delta = 1$ and $\gamma = 1 + \alpha$ we have $zE_{\alpha,1+\alpha}^1(z) = E_\alpha(z) - 1$. The extensive numerical calculation of $E_{\alpha,\gamma}(z)$ is considered in [36, 37], where the authors test the stability and the validity range for the parameters α and γ . Their algorithm is based on integral representations and exponential asymptotics. Using this algorithm they simulate the behavior of $E_{\alpha,\gamma}(z)$ also at zero and at infinity, and estimate the error of their method.

We attempt to express the generalized Mittag-Leffler function of Eq. (7) through a finite sum of the generalized hypergeometric function ${}_pF_q$ [30]. For this reason, we take the rational α , $0 < \alpha = l/k < 1$, and change the summation index as follows: $n \rightarrow kn + j$, where $n = 0, 1, \dots$ is the index which appears in Eq. (7). Here, we introduce the index $j = 0, 1, \dots, k-1$ which indicates how many generalized hypergeometric functions should be in the sum. Thus, we get

$$E_{l/k,\gamma}^\delta(z) = \sum_{j=0}^{k-1} \frac{z^j}{j!} \frac{(\delta)_j}{\Gamma(\gamma + \frac{l}{k}j)} \times {}_{1+k}F_{l+k} \left(\begin{matrix} 1, \Delta(k, \delta + j) \\ \Delta(k, 1 + j), \Delta(l, \gamma + \frac{l}{k}j) \end{matrix}; \frac{z^k}{l^l} \right) \quad (8)$$

with $\Delta(n, a) = \frac{a}{n}, \frac{a+1}{n}, \dots, \frac{a+n-1}{n}$. The first list of "upper" parameters is equal to 1 followed by $\Delta(k, \delta + j)$, whereas the second list of "lower" parameters is the union of $\Delta(k, 1 + j)$ and $\Delta(l, \gamma + \frac{l}{k}j)$. Let us now check if Eq. (8) reconstructs the exponential behavior of $E_{\alpha,\gamma}(z)$ used in [36, 37]. Starting from Eq. (8) and the last unnumbered formula on p. 155 of [38] we can estimate $E_{l/k,\gamma}^\delta(z)$ for large values of z . According to it ${}_{1+k}F_{l+k}$ function is equal to $Aj!\Gamma(\gamma + lj/k)/\Gamma(\delta + j)(k/l)^\delta \exp(z^{k/l})z^{k(\delta-\gamma)/l-j/k}$, where $A \rightarrow 1$ as $z \rightarrow \infty$, where we employed the Gauss-Legendre multiplication formula for gamma functions. Substituting it to Eq. (8) we have

$$E_{l/k,\gamma}^\delta(z) \simeq \frac{(k/l)^\delta}{\Gamma(\delta)} z^{k(\delta-\gamma)/l} e^{z^{k/l}}, \quad z \gg 1,$$

which for $\alpha = l/k$ and $\delta = 1$ reproduces the dominant term of [36, Eq. (2.4)] which presents the exponential behavior of $E_{l/k,\gamma}$.

Moreover, $E_{\alpha,\gamma}^\delta(z)$ for $\delta = \beta$ and $\gamma = \alpha\beta$ can be alternatively represented via the one-sided Lévy stable distribution $\Phi_\alpha(z)$ [15]:

$$E_{\alpha,\alpha\beta}^\beta(z) = \int_0^\infty e^{zs} \frac{s^{\beta-(1+\frac{1}{\alpha})}}{\Gamma(\beta)} \Phi_\alpha(s^{-\frac{1}{\alpha}}) ds. \quad (9)$$

Eq. (9) can be proved by writing the exponential function in the series form and using the explicit values of moments of $\Phi_\alpha(u)$, see [12, 15]. For closely related algebraic and transformation properties of Lévy stable functions consult [39–42]. Note that Eq. (9) generalizes the known relation between the classical Mittag-Leffler function and the one-sided Lévy stable distribution, see, e.g., [24, Eq. (7)] or [51, Eq. (11)]. Inserting the asymptotics of $\Phi_\alpha(u) \simeq u^{-1-\alpha}$ for large u given by [53, Eq. (5)] into Eq. (9) we get $E_{\alpha,\alpha\beta}^\beta(-(t/\tau_0))^\alpha \simeq (t/\tau_0)^{-\alpha-\alpha\beta}$. After substituting this result into Eq. (6) we reconstruct the asymptotics at infinity given in the last unnumbered formula on p. 76 of [33], namely $f_{\alpha,\beta}(t/\tau_0) \simeq (t/\tau_0)^{-1-\alpha}$. The behavior of $f_{\alpha,\beta}(t/\tau_0)$ for $t \gg 1$ looks like the asymptotics of $\Phi_\alpha(u)$ for $u \gg 1$. That suggests the link between the Prabhakar function and the one-sided Lévy stable distribution. We investigate this link in some detail in Sec. VI.

From Eqs. (6) and (8) with $\delta = \beta$ and $\gamma = 1 + \alpha\beta$ we write $f_{l/k,\beta}(t/\tau_0)$ as the finite sum of ${}_{1+k}F_{l+k}$'s functions. Moreover, $f_{l/k,\beta}(t/\tau_0)$ can be expressed through the standard special functions for $k = 2$ only:

$$f_{1/2,\beta}(\frac{t}{\tau_0}) = \frac{\sqrt{2}\tau_0\beta}{\sqrt{\pi}} \left(\frac{2t}{\tau_0}\right)^{\frac{\beta}{2}-1} e^{\frac{t}{2\tau_0}} D_{-1-\beta}(\sqrt{\frac{2t}{\tau_0}}), \quad (10)$$

where $D_\nu(z)$ is the parabolic cylinder function [45]. In the derivation of Eq. (10) we employ Eqs. (7.11.1.9) and (7.11.1.10) on p. 579 of [30].

To simplify the calculation in Sec. IV, we insert Eq. (9) with $\Phi_\alpha(z)$ given by [15, Eq. (2)] into Eq. (6) and employ formula (2.24.3.1) on p. 295 of [30]. Thus, the Prabhakar function can be expressed in terms of the Meijer G function $G_{p,q}^{m,n}$ [30]:

$$f_{l/k,\beta}(\frac{t}{\tau_0}) = (2\pi)^{\frac{1+l}{2}-k} \frac{\sqrt{l}k^\beta}{\Gamma(\beta)t} \times G_{l+k,k}^{k,k} \left(\frac{l^l\tau_0^l}{t^l} \left| \begin{matrix} \Delta(k, 1 - \beta), \Delta(l, 0) \\ \Delta(k, 0) \end{matrix} \right. \right). \quad (11)$$

The upper and lower lists of parameters are equal to the union of $\Delta(k, 1 - \beta), \Delta(l, 0)$ and $\Delta(k, 0)$, respectively. From Eq. (11) and Eq. (2.24.2.1) on p. 293 of [30] we calculate its μ -th Stieltjes moment $M_{l/k,\beta}(\mu) = \int_0^\infty t^\mu f_{l/k,\beta}(\frac{t}{\tau_0}) dt$ which has the form

$$M_{l/k,\beta}(\mu) = \tau_0^\mu \frac{\Gamma(\beta + \frac{k\mu}{l})}{\Gamma(\beta)} \tilde{M}_{l/k}(\mu), \quad (12)$$

where $\tilde{M}_{l/k}(\mu) = k\Gamma(-k\mu/l)/[\Gamma(-\mu)]$ denotes the Stieltjes moment of the one-sided Lévy stable distribution [15]. The Stieltjes moment $M_{l/k,\beta}(\mu)$ is finite for

$-l\beta/k < \mu < l/k$ and is infinite otherwise. Note that $\tilde{M}_{l/k}(\mu)$ is finite in the larger range of μ , namely for $-\infty < \mu < l/k$. $\tilde{M}_{l/k}(\mu)$ does not exist for $\mu \geq l/k$.

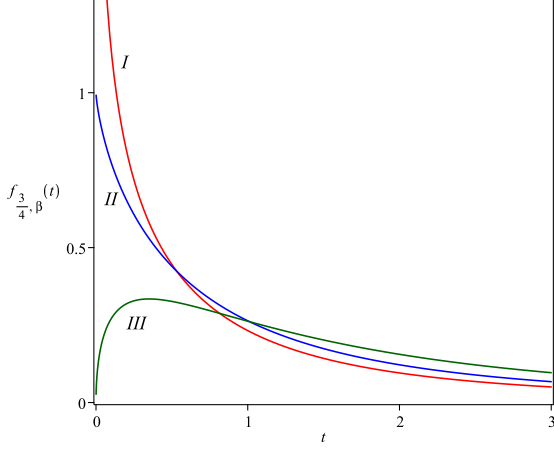


FIG. 1. (Color online) Plot of $f_{l/k, \beta}(t)$ given by Eq. (11) for $\tau_0 = 1$, $\alpha = 3/4$ and $\beta = 1$ (I; red), $\beta = 4/3$ (II; blue), and $\beta = 2$ (III; green).

Eq. (11) can be readily used to study the Prabhakar functions graphically. The Prabhakar functions $f_{3/4, \beta}(t)$ given by Eq. (11) for $\beta = 1, 3/4, 2$ are plotted in Fig. 1. In the limit of $t \rightarrow 0$ they go to infinity for $\beta > 4/3$, $f_{3/4, \beta}(t) \rightarrow 1$ for $\beta = 4/3$, and $f_{l/k, \beta}$ vanishes for $\beta < 4/3$. That exemplifies the asymptotic behavior of $f_{\alpha, \beta}(t) \propto t^{\alpha\beta-1}$ for $t \rightarrow 0$ [43] which goes to 1 for $\beta = 1/\alpha$. It also shows that $f_{3/4, \beta}(t)$ vanishes when t tends to infinity. This confirms the asymptotics found below Eq. (9).

IV. THE PROBABILITY DENSITY

The inverse Laplace transform allows one to pass from the time domain $t > 0$ to the space domain $u > 0$ and to address the question of emergence of probability distribution functions (pdf) in u .

The function $g_{\alpha, \beta}(u)$ is denoted here by $g(u)$. For $\alpha = l/k$, where l and k are integers such that $0 < l/k < 1$, $g_{l/k, \beta}(u)$ can be presented as the inverse Laplace transform of Eq. (5):

$$g_{l/k, \beta}(u) = \frac{\tau_0}{u} \mathcal{L}^{-1}[f_{l/k, \beta}(\frac{t}{\tau_0}), u]. \quad (13)$$

Substituting the Prabhakar function given by Eq. (11) into Eq. (13) and employing Eq. (3.38.1.2) on p. 393 of [44] we represent $g_{l/k, \beta}(u)$ in the form of the Meijer G function [30]:

$$g_{l/k, \beta}(u) = \frac{(2\pi)^{l-k} k^\beta}{\Gamma(\beta) u} G_{l+k, l+k}^{k, k} \left(u^l \left| \begin{matrix} \Delta(k, 1-\beta), \Delta(l, 0) \\ \Delta(k, 0), \Delta(l, 0) \end{matrix} \right. \right). \quad (14)$$

The symbol $\Delta(n, a)$ is defined after Eq. (8). The numerical tests show that $g_{l/k, \beta}(u)$ is a positive function for $0 < \beta \leq k/l$, whereas it has a negative part for $\beta > k/l$, see Fig. 2. (The analytical confirmation of that fact is below Eq. (22).) Employing Eq. (2.24.2.1) on

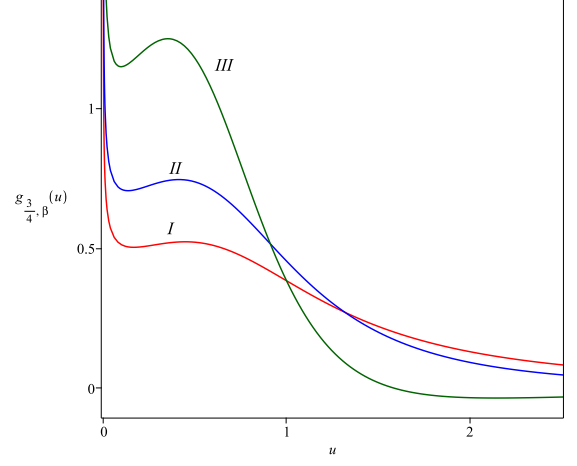


FIG. 2. (Color online) Plot of $g_{l/k, \beta}(u)$ given by Eq. (14) for $\alpha = 3/4$ and $\beta = 1$ (I; red), $\beta = 4/3$ (II; blue), and $\beta = 2$ (III; green). Observe that line III (green) is negative for $u > 1.33$.

p. 293 of [30] we show that all their fractional moments $N_{l/k, \beta}(\nu) = \int_0^\infty u^\nu g_{l/k, \beta}(u) du$ are equal to

$$N_{l/k, \beta}(\nu) = \frac{\Gamma(1 + \frac{k}{l}\nu) \Gamma(\beta - \frac{k}{l}\nu)}{\Gamma(\beta) \Gamma(1 + \nu) \Gamma(1 - \nu)}.$$

For real ν , $-\infty < \nu < l\beta/k$ and $\nu \neq -1, -2, \dots$, they are finite, and they are infinite otherwise. It means that $g_{l/k, \beta}(u)$ are normalized and their higher moments like the mean value or variance do not exist. Similar but not identical behavior is observed for the one-sided Lévy stable distributions $\Phi_\alpha(u)$ for which only fractional moments exist and all the higher moments are infinite [15]. The existence of normalization and positivity of $g_{l/k, \beta}(u)$ for $0 < \beta \leq k/l$ permits to conclude that $g_{l/k, \beta}(u)$ for $\beta < k/l$, k and l integers, are the pdf.

Furthermore, it turns out that using Eq. (8.2.2.4) on p. 617 of [30] and the Gauss-Legendre multiplication formula for gamma functions in Eq. (14), we can rewrite $g_{l/k, \beta}(u)$ as a finite sum of k generalized hypergeometric functions:

$$g_{l/k, \beta}(u) = \frac{1}{\pi} \sum_{j=0}^{k-1} \frac{(-1)^j}{j!} \frac{(\beta)_j}{u^{1+\frac{k}{l}(\beta+j)}} \sin[\frac{l}{k}(\beta+j)\pi] \times {}_{1+k}F_k \left(\begin{matrix} 1, \Delta(k, \beta+j) \\ \Delta(k, 1+j) \end{matrix}; \frac{(-1)^{l-k}}{u^l} \right). \quad (15)$$

Eq. (15) gives a closed form of $g_{l/k, \beta}(u)$. For example, for the CC relaxation ($\beta = 1$) it can be observed that the appropriate cancelation in ${}_{1+k}F_k$'s gives ${}_1F_0(\frac{1}{0}, (-1)^{l-k} u^{-l/k})$ which, after applying Eq. (7.3.1.1)

on p. 453 of [30], yields $u^l/[u^l - (-1)^{l-k}]$. Employing Eq. (1.353.1) on p. 38 of [45] to the remaining sum over j we get

$$g_{\alpha,1}(u) = \frac{u^{\alpha-1} \sin(\alpha\pi)}{\pi(u^{2\alpha} + 2u^\alpha \cos(\alpha\pi) + 1)}, \quad (16)$$

where $0 < \alpha = l/k < 1$. We point out that Eq. (16) was obtained in [46] by using a different method, see Eq. (3.24) on p. 245 there. All the distributions $g_{\alpha,1}(u)$ share the following features: (i) $g_{\alpha,1}(u)$ for $0 < \alpha < 1$ and $u \geq 0$ is positive; (ii) $g_{\alpha,1}(u)$ goes to infinity at $u = 0$, and it vanishes for $u \rightarrow \infty$; (iii) $g_{\alpha,1}(u)$ is an decreasing function for $\alpha < \alpha_0$, it contains the flat sector near $\alpha = \alpha_0$, and it has two extrema for $\alpha \geq \alpha_0$. For $\alpha = \alpha_0$ the derivative $g'_{\alpha,1}(u)$ is zero at the point $u = u_0$. The numerical calculation shows that for the CC relaxation we have $\alpha_0 \approx 0.737$ and $u_0 \approx 0.306$; (iv) $g_{\alpha,1}(u)$ has a maximum at $u = u_{\max}$ and a minimum at $u = u_{\min}$ for $\alpha \geq \alpha_0$, where $u_{\max/\min} = [-\cos(\alpha\pi) \pm (\alpha^2 - \sin^2(\alpha\pi))^{1/2}]/(1+\alpha)$. The upper sign is for maximum value of u and the lower sign is for minimum of u .

Eq. (15) offers an unlimited number of solutions for $g_{l/k,\beta}(u)$. For $k \leq 3$ only it can be written down in terms of standard special functions. For example, for $l/k = 1/2$ it is equal to

$$g_{\frac{1}{2},\beta}(u) = \frac{\sin(\beta \arctan(\sqrt{u}))}{\pi u(1+u)^{\beta/2}}, \quad (17)$$

which is plotted for $\beta = 3/4$ in Fig. 3, see line I in red, whereas $g_{l/k,\beta}(u)$ for $l/k = 1/3$ is equal to

$$g_{\frac{1}{3},\beta}(u) = \frac{(1 - u^{\frac{1}{3}} e^{\frac{2\pi i}{3}})^{-\beta}}{2\pi i u} - \frac{(1 - u^{\frac{1}{3}} e^{\frac{4\pi i}{3}})^{-\beta}}{2\pi i u}. \quad (18)$$

In the derivation of Eq. (17) we have used Eqs. (7.3.3.1) and (7.3.3.2) on p. 486 of [30]. Eq. (18) has been obtained by employing Eqs. (7.4.1.30) - (7.4.1.32) on p. 499 of [30]. From Eq. (17), after analyzing the behavior of sine, we conclude that $g_{1/2,\beta}(u)$ is positive for $0 < \beta \leq 2$, whereas it is negative for $\beta > 2$ and $u > [\tan(\pi/\beta)]^2$.

In Fig. 3 we exhibit the probability densities of $g_{l/k,\beta}(u)$ given by Eq. (15) for $\beta = 1/3$ and $\alpha = 1/2, 3/4, 19/20$. Fig. 3 shows that for fixed value of β there exists α_0 for which $g'_{l/k,\beta}(u)|_{u=u_0} = 0$. For example, for $\beta = 1/3$ it is $\alpha_0 \approx 0.747$ and $u_0 \approx 0.355$. For $\beta = 1/3$ and $\alpha > \alpha_0$ the maximum is higher and it moves in the direction of larger values of u .

We now find the series representation of $g_{\alpha,\beta}(u)$ for rational $\alpha = l/k$. To realize that objective we use the series representation of the generalized hypergeometric function, see Eq. (7.2.3.1) on p. 437 of [30]. In this way we obtain the double sum: one over r , $r = 0, 1, 2, \dots$, which is the index appearing in the series representation of ${}_pF_q$, and another one over j , $j = 0, 1, \dots, k-1$, is from Eq. (15). Next, we change the summation index as follows $kr + j \rightarrow r$. Thus, $g_{\alpha,\beta}(u)$ has the form

$$g_{\alpha,\beta}(u) = \frac{1}{\pi} \sum_{r=0}^{\infty} \frac{(-1)^r}{r!} \frac{(\beta)_r \sin[\alpha(\beta+r)\pi]}{u^{1+\alpha(\beta+r)}}. \quad (19)$$

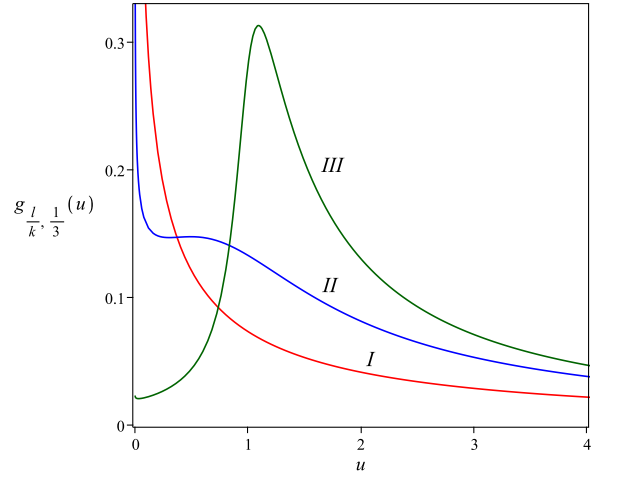


FIG. 3. (Color online) Plot of $g_{\alpha,\beta}(u)$ given by Eq. (15) for $\beta = 1/3$ and $\alpha = 1/2$ (I; red), $\alpha = 3/4$ (II; blue), and $\alpha = 19/20$ (III; green).

It turns out that writing sine as the imaginary part of $e^{i\pi\alpha(\beta+r)}$, taking the integral representation of the gamma function [45, Eq. (8.312.2)], and applying Eq. (7), we get the integral form of Eq. (19):

$$g_{\alpha,\beta}(u) = \frac{1}{\pi} \text{Im} \left\{ \int_0^\infty e^{-u\xi} G_{\alpha,\beta}(\xi e^{i\pi}) d\xi \right\}. \quad (20)$$

The auxiliary function $G_{\alpha,\beta}(x)$ written in terms of the (three-parameter) generalized Mittag-Leffler function is equal to

$$G_{\alpha,\beta}(x) = x^{\alpha\beta} E_{\alpha,1+\alpha\beta}^\beta(-x^\alpha). \quad (21)$$

Using Eq. (7) in Eq. (21) it is easy to demonstrate the following features of $G_{\alpha,\beta}(x)$. It can be shown that $G_{\alpha,\beta}(x)$ vanishes at $x = 0$. $G_{\alpha,\beta}(x)$ goes to 1 in the limit of $x \rightarrow \infty$. The asymptotics at $x \rightarrow \infty$ is presented in unnumbered formula for $\beta_M = \alpha\beta + 1$ and $\gamma_M = \beta$ on p. 76 of [33]. For the CC relaxation ($\beta = 1$), $G_{\alpha,\beta}(x)$ can be expressed via the classical Mittag-Leffler function, namely $G_{\alpha,1}(x) = 1 - E_\alpha(-x^\alpha)$ which is quoted in [24, Eq. (9) on p. 185]. For the CD relaxation ($\alpha = 1$) we have $G_{1,\beta}(x) = x^\beta E_{1,1+\beta}^\beta(-x) = 1 - \Gamma(\beta, x)/\Gamma(\beta)$ [47], where $\Gamma(\beta, x)$ is the incomplete gamma function. Moreover, $\frac{d}{dx} G_{\alpha,\beta}(x) = f_{\alpha,\beta}(x)$ which results from Eq. (1.9.6) on p. 46 of [35].

By applying [32, Eq. (2.5)] to Eq. (20) with Eq. (21) and by employing de Moivre's formula to calculate the imaginary part of so obtained equation, we derive the modification of $g_{\alpha,\beta}(u)$ which is given by Eq. (8) for $\alpha_M = \alpha$, $\beta_M = 1 + \alpha\beta$, and $\gamma_M = \beta$ of [33]. Namely, if $0 < \alpha < 1$ and $\beta > 0$ we get

$$g_{\alpha,\beta}(u) = \frac{1}{\pi u} \frac{\sin[\beta\theta_\alpha(u) + m\pi]}{[u^{2\alpha} + 2u^\alpha \cos(\pi\alpha) + 1]^{\beta/2}} \quad (22)$$

with $m = 0, 1$ (defining the range of u , see below), and

$$\theta_\alpha(u) = \arctan\left(\frac{u^\alpha \sin(\pi\alpha)}{u^\alpha \cos(\pi\alpha) + 1}\right) \in [0, \pi]. \quad (23)$$

For $\alpha \leq 1/2$ Eq. (22) works for $m = 0$ and $u > 0$. For $\alpha > 1/2$ Eq. (22) with $m = 0$ defines $g_{\alpha,\beta}(u)$ for $0 < u \leq [\cos(\pi\alpha + \pi)]^{1/\alpha}$. Eq. (22) with $m = 1$ gives $g_{\alpha,\beta}(u)$ for $u > [\cos(\pi\alpha + \pi)]^{1/\alpha}$. In this way we can define $g_{\alpha,\beta}(u)$ for all $u > 0$. Note that the presence of $m\pi$ in Eq. (22) allows us to build the positively defined and normalized pdf $g_{\alpha,\beta}(u)$ for arbitrary real α , $0 < \alpha < 1$ and $0 < \beta < 1/\alpha$. This property is absent in the solution of [33, Eq. (8)] for $\alpha_M = \alpha$, $\beta_M = 1 + \alpha\beta$, and $\gamma_M = \beta$, which is defined for $0 < \alpha < 1$ and $m = 1$. For example, $g_{3/4,1}(u) = K_{3/4,7/4}^1(u)$, where $K_{3/4,7/4}^1(u)$ is given by [33, Eq. (8)], is negative for $u \leq 1.587$. Introducing the term $m\pi$ we improve [33, Eq. (8)]. We remark also that Eqs. (14), (15), and (22) gives the same results.

Using the various but equivalent forms of $g_{\alpha,\beta}(u)$ make it easier to show many of its properties. For example, from Eqs. (22) and (23) it can be shown that $g_{\alpha,\beta}(u)$ is positive for $0 < \beta \leq 1/\alpha$, whereas it contains the negative parts for $u > [\tan(\pi/\beta)/(\sin(\alpha\pi) - \tan(\pi/\beta)\cos(\alpha\pi))]^{1/\alpha}$ for $\beta > 1/\alpha$. From Eq. (22), in a simple way, we get that $g_{\alpha,\beta}(u)$ goes to infinity in the limit of $u \rightarrow 0$ and $g_{\alpha,\beta}(u)$ vanishes for $u \rightarrow \infty$. The benefit of use Eqs. (14) or (15) rather than Eq. (22) is that it is the only formula which defines $g_{\alpha,\beta}(u)$ for $\alpha > 1/2$ and $u > 0$.

V. RELAXATION FUNCTION AND EVOLUTION EQUATION

We define now the relaxation function $n(t)/n_0 \equiv [n(t)/n_0]_{\alpha,\beta}$ whose explicit and exact form is obtained by calculating the Laplace transform (4) with $g_{\alpha,\beta}(u)$ given in Eq. (14). Here, we use the formulas from [30], i.e. Eq. (2.24.3.1) on p. 350, Eq. (8.2.2.14) on p. 619, and Eq. (8.2.2.3) on p. 618, and, then, we compare the results with Eqs. (21) and (8). Thus, we get [47, Eq. (30)]:

$$\left[\frac{n(t)}{n_0}\right]_{\alpha,\beta} = 1 - G_{\alpha,\beta}\left(\frac{t}{\tau_0}\right), \quad (24)$$

where $G_{\alpha,\beta}(t/\tau_0)$ is given in Eq. (21) with Eq. (8). Remark that Eq. (24) for $t = 0$ is equal to one and it vanishes for t going to infinity, see Fig. 4. Eq. (24) resembles the series representation of the relaxation function obtained in [28, 29]. It suggests that the HN relaxation could be explained by using the Debye's relaxation. For instance, for $\beta = 1$ the CC relaxation can be build from the Debye's relaxations [24]. In [28, 29] Eq. (24) is also represented in terms of the Fox H functions which however are not accessible in the computer algebra systems. The advantage of our solution, Eq. (24) with Eqs. (21) and (8), is clearly seen in practice. Since in recent

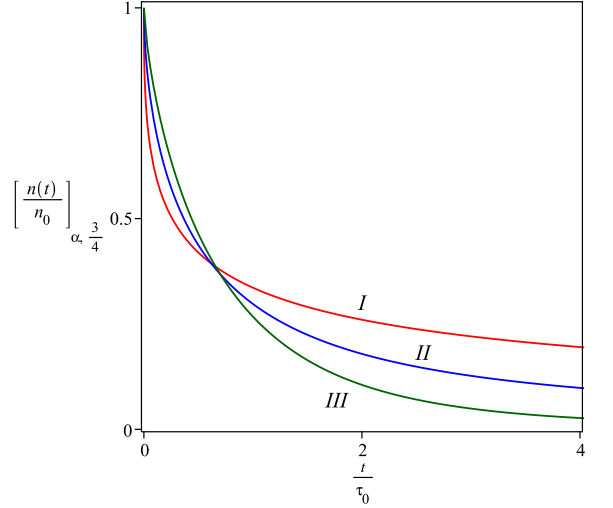


FIG. 4. (Color online) Plot of $[n(t)/n_0]_{\alpha,\beta}(x)$ given in Eqs. (24) and (8) for $\beta = 3/4$ and $\alpha = 1/2$ (I; red), $\alpha = 3/4$ (II; blue), and $\alpha = 19/20$ (III; green).

versions of the computer algebra systems the generalized hypergeometric functions ${}_pF_q$ as well as the Meijer G functions are fully implemented, their use permits high-precision calculations.

The explicit form of relaxation function $[n(t)/n_0]_{\alpha,\beta}$ given by Eq. (24) and Eq. (4) allow us to derive the self-similar properties of $g_{\alpha,\beta}(u)$. Rewriting the first equality in Eq. (4), where instead of t/τ_0 we take $a^{1/\alpha}p$, $a > 0$, and taking into account Eq. (24), we get

$$\begin{aligned} 1 - (ap^\alpha)^\beta E_{\alpha,1+\alpha\beta}^\beta(-ap^\alpha) &= \int_0^\infty e^{-pa^{1/\alpha}u} g_{\alpha,\beta}(u) du \\ &= \int_0^\infty e^{-px} a^{-1/\alpha} g_{\alpha,\beta}(a^{-1/\alpha}x) dx, \end{aligned}$$

where $u = a^{-1/\alpha}x$. An one-variable function $g_{\alpha,\beta}(u)$ is uniquely extended to a two-variable $\tilde{g}_{\alpha,\beta}(a, x)$ one:

$$\tilde{g}_{\alpha,\beta}(a, x) = a^{-1/\alpha} g_{\alpha,\beta}(xa^{-1/\alpha}). \quad (25)$$

We stress that Eq. (25) is the self-similar property which is also obeyed by the (classical) Mittag-Leffler function [35] and the Lévy stable distribution [15]. From the second equality in Eq. (4) with $a = (t/\tau_0)^\alpha$, $p = 1$, and for $t_0 \leq t_1 \leq t_2$ we get the Laplace-like convolution properties:

$$\begin{aligned} \int_0^x g_{\alpha,\beta}\left(\left(\frac{t_2-t_1}{\tau_0}\right)^\alpha, x-y\right) g_{\alpha,\beta}\left(\left(\frac{t_1-t_0}{\tau_0}\right)^\alpha, y\right) dy \\ = g_{\alpha,\beta}\left(\left(\frac{t_2-t_0}{\tau_0}\right)^\alpha, x\right). \end{aligned} \quad (26)$$

The similar property is fulfilled also by the one-sided Lévy stable distribution, see [16, Eq. (12)] and [48, Eq. (13)]. Eq. (26) differs from the standard Laplace convolution of *one* variable function. Here, we have the integral form of evolution equation of *two* variables density

distribution $g_{\alpha,\beta}((\frac{t}{\tau_0})^\alpha, x)$ where the *both* variables are changed. Eq. (26) can be proved by employing Eqs. (25) for $a = (t/\tau_0)^\alpha$, $p = 1$, and (16). Observe also that Eq. (26) is the evolution equation written in the integral form.

VI. $f_{\alpha,\beta}(t/\tau_0)$ AND $\Phi_\alpha(t)$

Following [24] the relation between HN relaxation and the one-sided Lévy density $\Phi_\alpha(u)$ is usually understood in the form of [24, Eq. (7)] or [51, Eq. (11)]. Here, we propose a new kind of link which will show the correlation of $f_{\alpha,\beta}(t/\tau_0)$, for arbitrary $0 < \alpha < 1$ and $\beta > 0$, with $\Phi_\alpha(t)$, $0 < \alpha < 1$. To achieve this task, we reparametrize in Eq. (1) $\beta = (2 - q)/(q - 1)$, $\tau_0 = (q - 1)^{1/\alpha}$ and $i\omega = \kappa$ throughout this Section. The range of parameter q , $1 < q < 2$, assures that $\beta > 0$.

The derivative of Eq. (1) over κ taken with the opposite sign is the probability density function

$$W_{\alpha,q}(\kappa) = \alpha(2 - q)\kappa^{\alpha-1}[1 - (1 - q)\kappa^\alpha]^{\frac{1}{1-q}} \quad (27)$$

called the q -Weibull distribution [49]. We point out that Eq. (27) goes to the Weibull distribution [49, 50] for $q \rightarrow 1+$ and the Weibull distribution is the derivative of the stretched exponential. Thereafter, we take the Prabhakar function given in Eq. (6) with the (three-parameter) generalized Mittag-Leffler function given via the finite sum of the generalized hypergeometric functions, see Eq. (8). Next, in Fig. 5 we compare

$$\tilde{f}_{\alpha,q}(t) \equiv f_{\alpha,(2-q)/(q-1)}(t(q-1)^{-1/\alpha}) \quad (28)$$

for $\alpha = 1/2$ and $q = 1.2, 1.1, 1.05$ with the $\Phi_{1/2}(t)$. In Fig. 5 we observe that $\tilde{f}_{1/2,q}(t)$ goes over to the one-sided Lévy stable distribution $\Phi_{1/2}(t)$ when q is approaching one.

The behavior of $\tilde{f}_{\alpha,q}(t)$ in the limit of $q \rightarrow 1+$ can be also shown by using in Eq. (6) and the series representation of the (three-parameter) generalized Mittag-Leffler function given in Eq. (7). After applying the Gauss-Legendre multiplication formula to gamma functions in the denominator of Eq. (7) we get

$$\begin{aligned} \tilde{f}_{\alpha,q}(t) &= \sum_{n=0}^{\infty} \frac{(-1)^n}{n!} \left(\frac{2-q}{q-1}\right)_n \left[\frac{t}{(q-1)^{-1/\alpha}}\right]^{\alpha(n+\frac{2-q}{q-1})-1} \\ &\times \Gamma[1 - \alpha(n + \frac{2-q}{q-1})] \sin[\alpha(n + \frac{2-q}{q-1})]. \end{aligned} \quad (29)$$

Writing the sine function as the imaginary part of $\exp[i\pi\alpha(n + \frac{2-q}{q-1})]$ and employing Eq. (8.312.2) on p. 892 of [45], the integral representation of $\tilde{f}_{\alpha,q}(t)$ is found in the form

$$\tilde{f}_{\alpha,q}(t) = \text{Im} \left\{ \int_0^\infty e^{-ty} [1 + (q-1)(ye^{-i\pi})^\alpha]^{-\frac{2-q}{q-1}} \frac{dy}{\pi} \right\}. \quad (30)$$

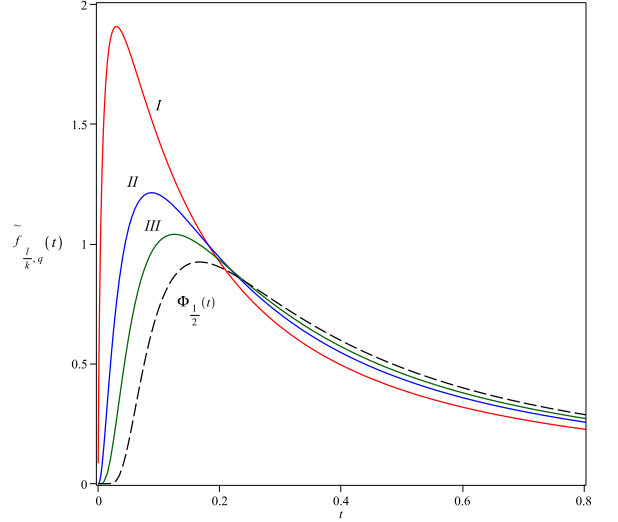


FIG. 5. (Color online) Plot of $\tilde{f}_{1/2,q}(t)$ given by Eq. (28), where the Prabhakar function Eq. (10) is defined for $\tau_0 = (q - 1)^{1/\alpha}$, $\beta = (2 - q)/(q - 1)$, and $q = 1.2$ (I; red), $q = 1.1$ (II; blue), and $q = 1.05$ (III; green). The black, dashed line shows the one-sided Lévy stable distribution for $\alpha = 1/2$, i.e. $\Phi_{1/2}(t) = \exp[-1/(4t)]/(2\sqrt{\pi}t^{3/2})$.

In the limit of $q \rightarrow 1+$ Eq. (30) goes to the one-sided Lévy stable distribution $\Phi_\alpha(t)$ given in the first unnumbered equation in the last page of [52].

The Stieltjes moments of $f_{\alpha,\beta}(t/\tau_0)$ given in Eq. (12) go to the Stieltjes moments of $\Phi_\alpha(t)$ for $q \rightarrow 1+$. To show that we express τ_0 and β through q , where $1 < q < 2$, namely by taking $\tau_0 = (q - 1)^{1/\alpha}$ and $\beta = (2 - q)/(q - 1)$. Thereafter, we apply the Stirling formula for $\Gamma(\beta + \nu/\alpha)$ and $\Gamma(\beta)$ where β is appropriately modified.

VII. CONCLUSION

Starting from the basic facts from relaxation theory we have introduced the response functions, the probability densities and the relaxation functions of the HN relaxation. The response functions are given via the Prabhakar functions related to the (three-parameter) generalized Mittag-Leffler functions whose representation in terms of the finite sum of the generalized hypergeometric functions for rational α is found. We also express the (three-parameter) generalized Mittag-Leffler functions via the one-sided Lévy stable distributions. That relation for $\beta = 1$ (the CC relaxation) goes over to the known relation between the (classical) Mittag-Leffler function and the one-sided Lévy stable distribution. In this way it generalizes the latter mentioned relation. We have also identified the values of parameters β , i.e. $0 < \beta \leq 1/\alpha$, for which the normalized function $g_{\alpha,\beta}(u)$ connected with the response function can be called the probability density. The moments of $g_{\alpha,\beta}(u)$ were also calculated. For $0 < \beta \leq 1/\alpha$ the mean values, the vari-

ance and the higher moments of $g_{\alpha,\beta}(u)$ do not exist. Similar properties characterize the one-sided Lévy stable distribution. We also derived the Laplace-like convolution properties which are the integral form of evolution equations. Moreover, we present the evidence that $f_{\alpha,\beta}(t/\tau_0)$ and the appropriate $\Phi_\alpha(t)$ present the identical asymptotic behavior at infinity. This suggests the existence of the link between $f_{\alpha,\beta}(t/\tau_0)$ and $\Phi_\alpha(t)$. This link has been also considered.

The main benefit of the paper is that, in addition to finding the explicit and exact forms of the relaxation functions, we represent them as the generalized hypergeometric functions which are implemented in the computer algebra systems. The use of the generalized hypergeometric functions have significantly simplified the calculation and in the fast way allowed us to obtain many results. Another advantage is that we found restrictions on β for which $g_{\alpha,\beta}(u)$ is the probability density. For this value of β , $g_{\alpha,\beta}(u)$ gives localization of relaxing centers in

the sample. That could give the intuition in which way we should prepare the experimentally measured sample. We have also elucidated the way in which the various versions of $f_{\alpha,\beta}(t/\tau_0)$, $g_{\alpha,\beta}(u)$, and $[n(t)/n_0]_{\alpha,\beta}$ appearing in the literature can be connected with each other. We have also shown the existence of new relations between the response function related to the HN relaxation, and the one-sided Lévy stable distribution, which can allow us to find the differential equation related to the integral Laplace-like convolution.

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